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OCCURRENCE AND RISK MANAGEMENT OF MAIN TOXIC ELEMENTS IN FOOD PRODUCTION CHAIN

Jasna S. Mastilović[1], Žarko S. Kevrešan[1], Elizabet P. Janić-Hajnal[1], Aleksandra R. Novaković[1], Tanja I. Radusin[1]

[1]Institute for Food Technology, University of Novi Sad,
Bul. cara Lazara 1, Novi Sad, Serbia

ABSTRACT: On the basis of available multiannual results of testing of the most common toxic metals in agricultural and food products in Serbia and in EU, the preliminary evaluation of risk of exposure of consumers in Serbia to health risks and incidents that can be provoked by intake of food containing toxic metals was undertaken. Based on information concerning the paths of introduction of toxic metals into food production chain the critical points of entrance of toxic metals into the food production chain were identified. The results indicate that risk of introduction of toxic metals into the food chain through agricultural products can be rated as low while the risk of introduction of toxic metals through spices, additives, aromas and supplements is higher. The conducted analysis indicates that exposure of consumers in Serbia to toxic metals as food contaminants is at quite low level.

Keywords: *toxic metals, risk management, food contamination*

INTRODUCTION

Toxic elements in food are defined as elements from Mendeleev periodic table which can be present in food in amounts that can be potentially hazardous to human health. Toxic elements in food originate from several different sources among which the most frequently studied are agricultural production of plants and animals, industrial or household processing and migration from package during storage of final products (Deshpande, 2002).

Although all elements can interact with human body it is possible to differentiate between elements which are considered to be essential and the ones that cause toxicological symptoms at extremely low levels and have no known beneficial physiological functions. The main toxic elements that are representative of the second group are mercury, lead, cadmium and arsenic. Mentioned toxic elements could contaminate agricultural soils, water supplies and environment and consequently found their way into the human food chain (McLaughlin et al., 1999).

The predominant route of entrance of toxic elements into the food chain arises during agricultural plant cultivation, through absorption of macro- and micronutrients from the soil by plants. Soils can naturally be rich in some highly toxic elements. Some of the contaminants, especially soluble ones, are present in soil solution in high concentration. For example, cadmium content in soils originating from crystal rocks was determined to vary from 1 to 90000 μg kg^{-1} (Page et al., 1981; Cook and Freny, 1988; Kabata-Pendias and Pendias, 1992) and for the soils that overlay some sedimentary

*Corresponding author:
E-mail address: jasna.mastilovic@fins.uns.ac.rs
Phone: +381214853773
Fax: +38121450725

rocks the content of several hundreds of mg of As per kg of soil was reported (Peterson et al., 1981). Besides natural occurrence, elevated concentrations of inorganic contaminants in soil can also be a result of human activities. It was established that elevated concentrations of arsenic in soil can be the consequence of coal burning in power stations (Wickstrom, 1982) while cadmium contamination of rice in Japan was linked to the contaminated fields (Asami, 1991). Soils can also be polluted by other industrial activities such as ore melting or battery production. Abundant traffic is significant source of soil pollution with lead (Chow, 1970). As one of the most studied pollutants, it was tracked that lead reaches food chain as a result of combustion of lead containing petrol additives, smelting and other anthropogenic activities (Alloway, 1990). Regardless the route by which lead finds its way to the soil, it mainly occurs in upper layers of soil profiles due to its intensive sorption by soil (Cartwright et al., 1977; Gulson et al., 1981; Merry et al., 1983). Having in mind anthropogenic sources of environmental pollution, intensive agriculture itself can significantly contribute to soil pollution. For example arsenic is used as a constituent of some insecticides, herbicides and defoliants (McLaughlin et al., 1999) while mercury is introduced into the soil as a constituent of fungicides, fertilizers, limestone, natural gypsum and manure (Andersson, 1979). Avoidance of soil pollution by some inorganic contaminants, for example cadmium, is somewhat impossible to achieve. Namely, cadmium is present practically in all phosphate fertilizers as an impurity (McLaughlin et al., 1999).

Availability of inorganic contaminants in food plants from soil depends on many factors, including soil properties, e.g. pH value, amount of clay, amount and type of organic compound in soil, redox potential, textural class of soil, and the presence of microorganisms. By influencing the factors which determine the availability, agricultural practice can affect the available/unavailable ratio of toxic metals (McLaughlin et al., 1999).

Uptake of toxic metals by terrestrial plants is often considered as entering point of many inorganic food contaminants in food chain and thus is the subject of many studies oriented to different metals, concentrations, species, genotypes and translocation rates (Kevrešan and Petrović, 2003; Petrović et al., 1998). Alternatively, toxic elements can enter the food chain from different environmental compartments, for example from fresh water plants that are also able to uptake the toxic metals from rivers and canals (Pajević et al., 2002), or through food animals via feed additives containing dangerous elements like e.g. arsenic. Small, localized incidents occurring quite frequently, for example by consuming the seeds treated with mercuric fungicides (Bakir at al., 1973), can also be considered as entering point of inorganic contaminants into the food chain.

In the case when inorganic contaminants are taken up by food plants from soil the main portion of toxic elements remain in root and plant organs that were in direct contact with the soil. On the other hand, depending on the plant type, fruits can be the most important edible parts of plants and thus when considering the contamination of food plants by inorganic contaminants, the focus has to be on the transport of inorganic contaminants to edible parts of plants. Some contaminants such as arsenic are toxic to plants at low concentrations and their accumulation in roots causes plant injury preventing further transport to aboveground parts (Peterson et al., 1981; O'Neill, 1990).

Cadmium concentration in plant food can vary widely. In general, the highest concentrations of cadmium are present in roots followed by steams and leaf and the lowest amounts are registered in fruits and seeds (Kevrešan et al., 2004). In this respect oilseeds are exception containing high levels of cadmium and thus being often regulated individually concerning cadmium content.

In animal food products one of the most important factors that influence the content of toxic metals in animal products is the life span of the animal. Animals with long life span such as horses, older cattle and game, can accumulate some inorganic contaminants. When exposed to feed contaminated with toxic metals, long-lived animals accumulate metals in blood, bones and meat. The edible organs where the highest concentrations of metals are expected are kidneys and liver (Janković et al., 2008; Sharma et al., 1982). Despite the fact that game usually is not a part of regular daily diet it is important to mention that it can be contaminated with lead, cadmium and other contaminants (Janković et al., 2008; Tojagić et al., 2004). Considering toxic metal content meat from intensively produced animals, especially ones with short life span, represents low risk (Pavlović et al., 2001; Janković et al., 2008).

As claimed by Marvin et al. (2009) there is a need for early identification of food safety issues in order to prevent them from developing into health risks. Various methods and procedures which can be used for early identification of safety issues including the monitoring of the occurrence of specific hazards within the food supply, or the incidences of food-borne diseases, as well as the combination of these data with other data or with expert opinions, enable pro-active operating by pre-defining indicators for hazards and follow-up measures based on HACCP principles.

The identification of safety issues in Serbia is by the rule performed on the basis of testing of regulatory prescribed contaminants, including mentioned toxic metals, in samples submitted by entities participating in food production chain to chosen authorized laboratory. Neither of above mentioned methods and procedures of systematic food safety monitoring process was up to now applied in Serbia, resulting in non existence of evaluation of food safety risks to which consumers in Serbia are exposed to.

The aim of this paper was to provide, on the basis of available multiannual results of testing of the most common toxic metals in agricultural and food products in Serbia and EU, the preliminary evaluation of risk of exposure of consumers in Serbia to health risks and incidents that can be provoked by intake of food containing toxic metals.

MATERIAL AND METHODS

Results of reported contamination of agricultural and food products with toxic metals were used for the same three-year period from two different sources:

a) From data base of accredited Laboratory for Food Technology, Quality and safety in the Institute for Food Technology in Novi Sad, Serbia in which method of atomic apsortion spectroscopy is used for the determination of toxic metals (Cd, Pb were determined with Varian SpectrAA 10 using flame technique while for As $NaBH_4$ technique were used. For the determination of Hg dedicated AAS instrument (Leco AMA254) for direct Hg determination was used).

b) From EU Rapid Alert System for Food and Feed data base portal (RAFFS) in which all incidents resulting in withdrawal of food or feed from the market, in recommendations for preventing, limiting or imposing specific conditions on the placing on the market or the eventual use of food or feed on account of a serious risk to human health or in rejection on EU border posts are notified (https://webgate.ec.europa.eu/rasff-window/portal/)

The results were statistically processed and frequencies of samples, in which incidence of levels of toxic metals nonconforming with regulations in force was detected, were determined. The samples were categorized according to:

a) Their position in food production chain for results obtained in Serbia (agricultural products, food ingredients and food products)

b) The food products category to which they belong for results taken over from RAFFS data base portal.

The results were utilized for evaluation of risk of incidence of toxic metals in Serbia in critical points determined along the food production chain. The critical points for occurrence of toxic metals were indicated on the basis of provided literature review.

RESULTS AND DISCUSSION

Based on the information concerning the paths of introduction of toxic metals into food production chain it can be summarized that toxic metals enter the food production chain, as presented in figure 1:

a) Through agricultural products utilized as raw materials;
b) Through other ingredients utilized in food production;
c) From equipment utilized in warehousing and processing operation;
d) Through preparations utilized in sanitation and pest control processes:
e) From packaging materials during storage of final products.

Toxic metals may be introduced into agricultural plant products used as raw materials in food production from soil, fertilizers and pesticides, deposits and irrigation water. Utilization of contaminated plant products, as feed constituents, presents the pathway of introduction of toxic metals into animal products. Starting contamination with toxic metals introduced into food production chain through utilized agricultural raw materials cannot be removed in the processes of food production but it is, depending on applied technological processes and operations, being diluted and concentrated resulting in final level of toxic metals in final food products. Risk management in agricultural production is the only efficient way of reduction of occurrence of toxic metals originating from agricultural raw materials. Poor risk management in agricultural production leads to the demand of permanent expensive and extensive control of level of toxic metals in agricultural raw materials at their entrance in food production.

All ingredients utilized in food production deliver the original toxic material content into the food product for which they are used. Depending on composition of food products the content brought into food products through ingredients is being diluted to the level related to share of every ingredient in final product. Management of contamination with toxic metals from food ingredients can be conducted through:

- Reliance on risk management processes in ingredients production or
- Control of level of toxic metals present in ingredients.

Sources of contamination with toxic metals positioned in warehousing, processing and packaging operations can be equipment, pest and sanitary control preparations and packaging materials. Absence of risk management in this part of food production chain including certification of equipment and materials which it is made of, testing and certification of packaging materials related to duration of storage of packaged food products and composition of stored product and prevention of entrance of sanitation and pest control preparations in food chain, can be detected only through control of toxic metal levels in every lot of final food products.

Once the food product is finalized and packed in safe packaging material, no further introduction of toxic metals and no decrease of present toxic metals in food products can occur.

Incorporation of Hazard Analysis Critical Con-trol Points (HACCP) allows the assessment of the risk and estimation of hazards severity, which may be associated with the used raw materials, their processing and the distribution system. HACCP plans ensure that product safety is sustainable (Baker, 1995). According to Notermans et al. (1995) an important aspect of the hazard analysis critical control point (HACCP) system is the specification of criteria with setting the critical limits which ensure that the activities at a specified critical control point (CCP) are under control.

As presented in Figure 1, apart from border control of level of toxic metals present in imported food products, the critical control points have to cover the raw material from agricultural production and the ingredients utilized in food production.

The results presenting frequency of incidence of toxic metals in agricultural raw materials and food ingredients with list of products in which the contaminations were detected in the observed three year period are shown in Table 1.

Results indicate that lowest percent of detected metals is in agricultural raw materials. Lead, mercury and arsenic were not detected in any of 82 tested products while cadmium was detected only in one, imported agriculture product (peanut) leading to the conclusions that the risk of introduction of toxic metals into food production chain in Serbia through agricultural raw materials is very low.

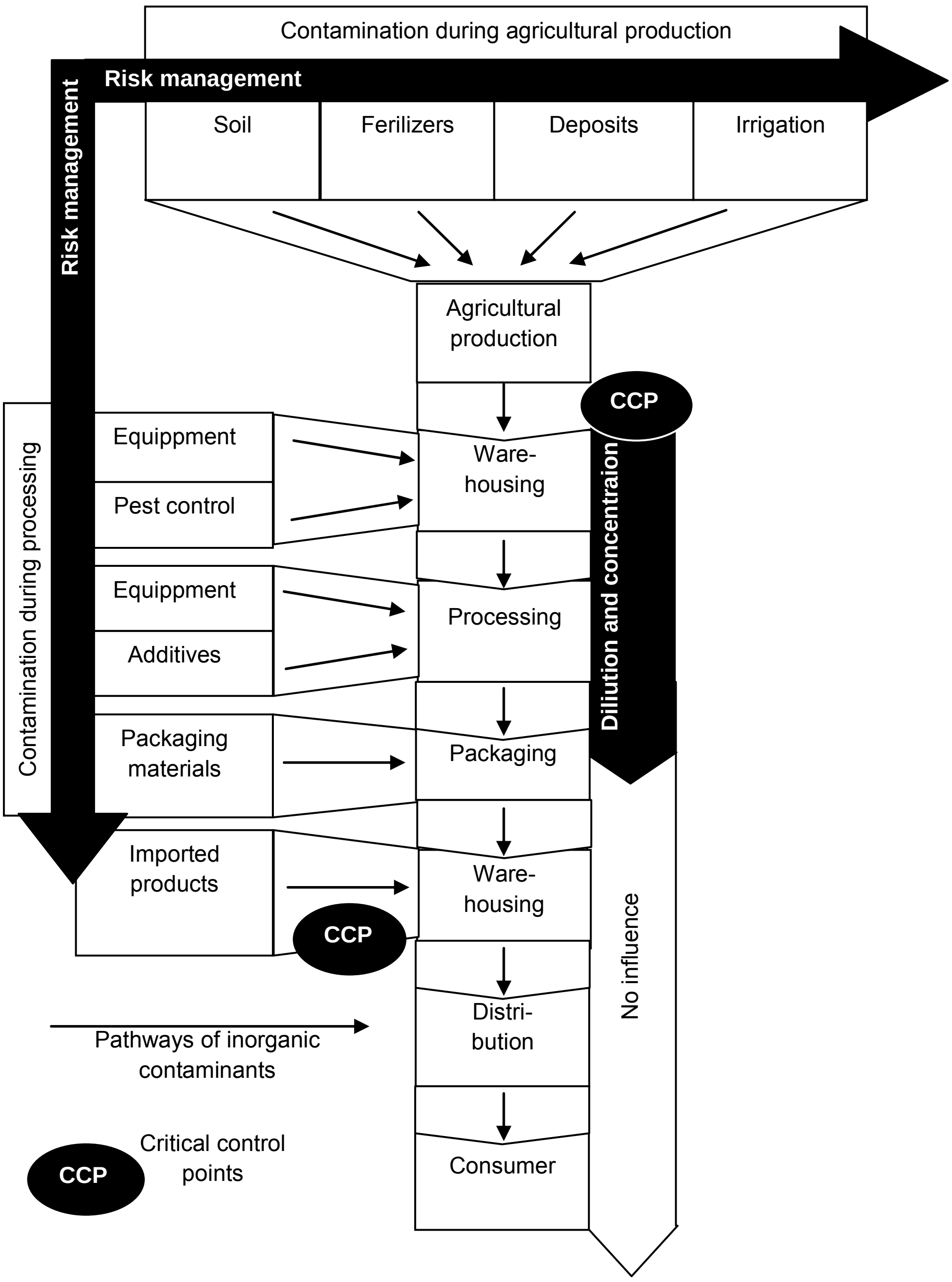

Figure 1. Possible paths of entering of toxic metals into the food chain and the control points

Incidence of toxic metals in ingredients utilized in food production is slightly higher. Presence of toxic metals, nevertheless in quantities lower than MRL (maximum residue limit) defined in official regulations, was determined in less than 2% of tested samples. The most frequent contaminant detected was lead detected to be present in 1,68% of tested products including mixtures of spices, tea, barbeque sauce, isolated soy protein followed by cadmium detected in 1,53% of tested samples including mixtures of spices, wheat flour, additive mixture, poppy staffing, potato starch and feed meal.

Arsenic and mercury were determined in approximately 0,5% of tested samples represented with samples of buckwheat flour, corn grits, in which arsenic was detected and additive, aroma, in which mercury was detected. Having in mind that ingredients in the food production are further diluted due to their low shares in final food products composition the risk of contamination of food products through food ingredients can also be categorized as low.

Table 1.
Frequency of incidence of toxic metals (lead, cadmium, arsenic, mercury) in agricultural raw material and food ingredient samples from Serbia

	No of samples (total)	Number of samples in which toxic metals were not detected	% of samples in which metals were detected	Number of samples in which toxic metals were detected (lower than MRL*)	Number of samples in which toxic metals were detected (higher than MRL*)
Agricultural products					
Pb	84	84	0,00	0	0
Cd	81	80	1,25	1(1)	0
As	84	84	0,00	0	0
Hg	81	81	0,00	0	0
Ingredients for food production					
Pb	482	476	1,26	8(2)	0
Cd	399	393	1,53	6(3)	0
As	477	475	0,42	2(4)	0
Hg	364	362	0,55	2(5)	0

(1) Products: Peanut
(2) Products: mixtures of spices, tea, barbeque sauce, isolated soy protein
(3) Products: spice mixtures, wheat flour, additive mixtures, poppy staffing, potato starch, feed meal
(4) Products: buckwheat flour, corn grits
(5) Products: additive, aroma
*MRL: maximum residue limit

Table 2.
Frequency of incidence of toxic metals (lead, cadmium, arsenic, mercury) in food products in Serbia

	No of samples (total)	Number of samples in which toxic metals were not detected	% of samples in which metals were detected	Number of samples in which toxic metals were detected (lower than MRL*)	Number of samples in which toxic metals were detected (higher than MRL*)
Food Products					
Pb	712	673	5,79	39(1)	0
Cd	660	654	0,92	6(2)	2(3)
As	711	710	0,14	1(4)	2(5)
Hg	657	650	1,08	7(6)	0

(1) Products: fruit and vegetable, pulses, chestnut puree, vinegar, bran, salad dressing
(2) Products: bran,
(3) Products: roasted wheat germ,
(4) Products: cookies with sesame seed
(5) Products: water (springs)
(6) Products: radish, lemon, onion, mushrooms
*MRL: maximum residue limit

The most frequent contaminant from the group of toxic metals that could be detected in food products from Serbian market is lead which was detected at levels under the MRL primarily in fruit and vegetables but also in samples of chestnut puree, vinegar, wheat bran and salad dressing.

Cadmium is determined to be the most frequent contaminants of wheat milling products with high nutritive value: wheat bran for human nutrition in which cadmium is often detected at levels lower than MRL and roasted wheat germ in which cadmium was detected to be present at the levels above MRL. This result indicates that cadmium in wheat milling products has to be analyzed more thoroughly concerning its paths of entrance into food chain but also concerning the risk that it represents to the consumers nutrition.

Out of more than 700 samples only one incident of food contamination with arsenic determined in cookies with sesame seed was detected in quantities under MRL. Quantities of arsenic under MRL were detected only in spring water which was due to this finding forbidden for utilization.

There were no products detected in which mercury level was determined to be above MRL. Levels of mercury under MRL were detected in different fruit, vegetable and mushroom products indicating another point in which more careful risk analysis has to be carried out.

Table 3.
Number of food products by categories in which toxic metal (lead, cadmium, arsenic, mercury) risk was reported in EU

Category	Cd	Pb	Hg	As
sea food	142(1)	7(2)	307(2)	0
fruit and vegetables	8(4)	14(5)	0	14(6)
meat and meat products (other than poultry)	5(7)	1(8)	0	0
cereals and bakery products	4(9)	1(10)	0	0
cocoa and cocoa preparations, coffee and tea	0	11(11)	0	1(12)
nuts, nut products and seeds	0	2(13)	0	0
non-alcoholic beverages	0	1(14)	0	0
dietetic foods, food supplements, fortified foods	1(15)	14(16)	3(17)	12(18)

Countries of origin: Sea food:	(1-Cd) France, UK, Greece, Vietnam, India, Indonesia, Thailand, China, Greece, Argentina, Australia, Denmark, Italy, Marocco, Chile, Philipines, Oman (2-Pb) Greece, Taiwan, South Africa, Senegal (3-Hg) Spain, Denmark, Portugal, Namibia, Italy, Uruguay, Cyprus, Senegal, Sri Lanka, Mauritius, Indonesia, South Africa, Singapore, Ghana, Vietnam, Algeria, China, Vietnam, Yemen, Morocco, Colombia, Brazil, Panama, Netherlands, New Zealand, France, Tunisia, US, Uruguay, Taiwan, Ecuador, UK, Croatia, Ivory Coast, Chile, Fiji, Tanzania, Algeria, Japan, Uganda, India, Seychelles, Madagascar
Fruit and vegetable:	(4-Cd) Spain, China, Belgium, Netherlands, Italy (5-Pb) Netherlands, Ukraine, Moldova, Pakistan, Canada, China, Spain (6-As) Japan, Hong Kong, China, United States, Korea,
Meat and meat pr. :	(7-Cd) Poland, Italy, Austria, Bulgaria, Mexico, China, Germany (8-Pb) Italy
Cereals and bakery pr.:	(9-Cd)Many countries but raw material from China (10-Pb) raw material for bakeries from China
Cocoa, coffee and tea:	(11-Pb) China, Belgium, France, Germany (12-As) China
Nuts and seeds:	(13-Pb)Moldova, Ukraine
Beverages:	(14-Pb) Iran
Dietetic foods:	(15-Cd) Switzerland (16-Pb) France, United States, Switzerland, India, Nigeria, Austria, Netherlands, Austria, Germany, China (17-Hg) United Kingdom, India (18-As) India, Netherlands, Austria (from China), France, Canada, United States, France (from Netherlands), Austria

In order to obtain insight into the possible sources of toxic metals contamination in food samples, the number of food products by food categories reported in EU Rapid Alert System for Food and Feed data base portal notified to bear the risk of toxic metals, is presented in table 3. The indication of country of origin of foodstuffs contaminated with toxic metals is also provided under the table.

The main source of toxic metals in diet of EU citizens is sea food. Independently of region of origin the frequency of contamination of sea food cadmium and especially with mercury is high above any other food product. Having in mind that sea food, due to its high price, is quite rare product in the diet of consumers in Serbia it can be concluded that this risk is in Serbia of lower expected impact on health status of population.

Out of frequently consumed products the fruits and vegetables can be indicated as the most frequent sources of contamination with lead, arsenic and even cadmium. These contaminants were detected in differrent fruits and vegetables originating equally from Far East countries, countries of east Europe or EU and North American countries. Having in mind the pathways of entrance of toxic metals into food chain through fertilizers, pesticides, deposits, soil and air contaminants this result is not surprising.

Meat and meat products from different countries and regions were detected to be rarely contaminated with cadmium and even more rarely with lead but nevertheless, this risk has also to be considered. Cadmium and lead were also detected to be sometimes present in cereals and bakery products. It is important to notice that in all identified cases raw material originated from China indicating it as the region of higher risk of occurrence of cadmium and lead contamination in raw material for bakery products.

Cacao and cacao products are quite often contaminated with lead indicating the need for consideration of lead as cacao based products contaminant more carefully.

Contamination of nuts and nut products and non alcoholic beverages with lead can be considered as incidents which do not represent rule, but indicates the need for interventions at the level of particular producers.

Special emphasis should be given to dietetic products which are quite often contaminated with lead and arsenic but carry sometimes also the risk of contamination with cadmium and mercury. When considering this group of products the fact that the prescribed levels of tolerance in dietetic and fortified products is lower than in ordinary products of the same type.

CONCLUSIONS

Although the risk management in agricultural production in Serbia is quite poor, occurrence of toxic metals in agricultural raw materials can be rated as rare. Main sources of contamination of food with toxic metals in Serbia are spices, additives, aromas and supplements but due to their dilution in production of final food products the risk for the consumers can be rated as not very high.

Exposure of consumers in Serbia to the risk of intake of toxic metals through food based on presented results is also not high. Detected levels of lead in fruit and vegetables, cadmium in wheat milling products and mercury in vegetables indicate these points as critical for more thorough risk analysis.

Among imported products, based on EU experience, sea food in the first instance, but also fruits and vegetables, cacao and cacao products, dietetic and fortified products and raw materials for bakery products from China should be considered as main carriers of contamination with certain toxic metals.

The main conclusion concerning chemically incorporated inorganic contamination is that there are no procedures or processing operations for the removal of these risks from the product once they have been brought into the production chain either in the form of contaminated agricultural products, additional raw materials, additives, and packaging materials, or as the contamination from equipment or used chemicals. Risk management of possible contamination sources in agricultural production and processing on one side, and prevention of entering of contamination into the food production chain on the other side, anticipates the definition of the critical control points wherever significant quantities of new materials are introduced into the food production chain. Food processing procedures result in dilution of contaminant concentration, when contaminated ingredient is only part of the final product (e.g. wheat germ as ingredient in fine bakery products), or its concentration, when the most contaminated part of starting raw material becomes the final product.

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